

REPORT DOCUMENTATION PAGE

AFRL-SR-BL-TR-98

Public report no burden for this collection of information is estimated to average 1 hour per response, gathering and maintaining the data needed, and completing and reviewing the collection of information, collection of information, including suggestions for reducing this burden, to Washington Headquarters Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Project of this 5 Jefferson

02135

1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE 4/24/98	3. REPORT TYPE AND DATES COVERED Final 01 Jun 94-30 Nov 97
4. TITLE AND SUBTITLE Novel Quantum Dot-Waveguide Devices by the Sol-Gel Method			5. FUNDING NUMBERS F49620-94-1-0343 1651/01 62173C
6. AUTHOR(S) N. Peyghambarian, E Wright, S. Honkanen, P. Guerreiro			8. PERFORMING ORGANIZATION REPORT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) The University of Arizona Optical Sciences Center Tucson, AZ 85721			10. SPONSORING / MONITORING AGENCY REPORT NUMBER
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR/NL 110 Duncan Avenue, Room B115 Bolling AFB, DC 20332-8050			
11. SUPPLEMENTARY NOTES			
12a. DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution unlimited.			12b. DISTRIBUTION CODE
13. ABSTRACT (Maximum 200 words) Passive CW mode-locking of a Cr:forsterite laser using PbS quantum-dot glass samples as intracavity saturable absorbers was demonstrated. This is believed to be the first application of a quantum-dot system in a practical device. Average output powers of 74 mW, 4.6 ps laser pulses at 110 MHz repetition rate, and a wide tunability range of 1207 to 1307 nm were obtained. The modelocking was made possible by the absorption saturation in the PbS quantum-dot samples, observed at room temperature. For the first time, significant optical gain was measured in quantum-dots pumped with nanosecond-pulse excitation. The samples studied were sol-gel derived CdS quantum-dots. The gain persists up to room temperature, it has a broad spectral width, and has its maximum value slightly below the absorption band edge. The measured rather surprising gain features were confirmed by theoretical calculations. Ultrashort pulse propagation in quantum dot waveguides was investigated. The measured characteristics of femtosecond pulse propagation, near two-photon resonance, in a CdS quantum-dot waveguide were theoretically explained. The new theoretical models developed open the doorway to propagation of space-time solitons in quantum-dot waveguides.			
14. SUBJECT TERMS Quantum-Dot, Sol-Gel, Modelocking, Saturable Absorber, Quantum-Dot Waveguide, Soliton			15. NUMBER OF PAGES 4 16. PRICE CODE
17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT

Novel Quantum Dot-Waveguide Devices by the Sol-Gel Method

N. Peyghambarian, E. Wright, P. Guerreiro and S. Honkanen
Optical Sciences Center, University of Arizona, Tucson, AZ 85721

We have demonstrated passive mode-locking of a Cr:forsterite laser using our PbS quantum-dot (QD) glass samples as intracavity saturable absorbers.¹ **We believe that this is the first application of a quantum-dot system in a practical device.** To use the PbS QD samples as saturable absorbers in the laser cavity, we polished PbS-doped glass plates down to approximately 150 μm for adequate absorption. At this thickness the intracavity losses were low enough to obtain reasonable output power and the saturable absorption was sufficient for mode locking. The sample was then anti-reflection coated with a quarter-wave layer of MgF_2 for 1250 nm to minimize Fresnel reflection losses. At this wavelength the thinned sample had a transmittance of 96%. Then the sample was placed in contact with the flat 2% output coupler of the laser (see Fig 1). Finally this glass with PbS quantum dots was used as an intracavity saturable absorber in a Cr:forsterite laser, resulting in the passive CW mode locking of the laser. We obtained average output powers of 74 mW, 4.6 ps laser pulses at 110 MHz repetition rate, and a wide tunability range of 1207 to 1307 nm (see Fig 1 and 3).

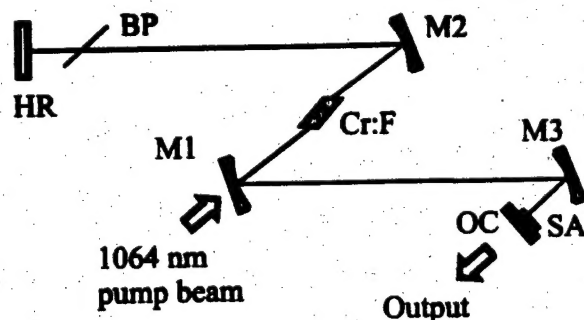


FIG. 1 Schematic of the laser cavity: Cr:F-19 nm Cr:forsterite crystal; M1-M3-concave mirrors ($R=10$ cm); HR-high reflector; OC-2% output coupler; SA-PbS quantum-dot glass saturable absorber; BP-birefringent plate used to tune the laser.

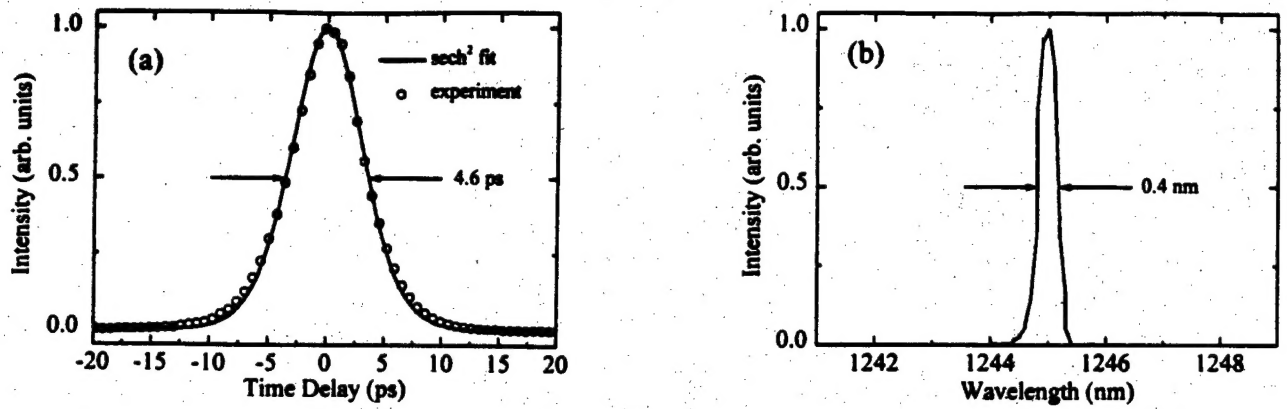


FIG. 2 (a) Intensity autocorrelation and (b) spectrum of near transform limited mode-locked pulses from the Cr:forsterite laser with PbS QD doped glass saturable absorber.

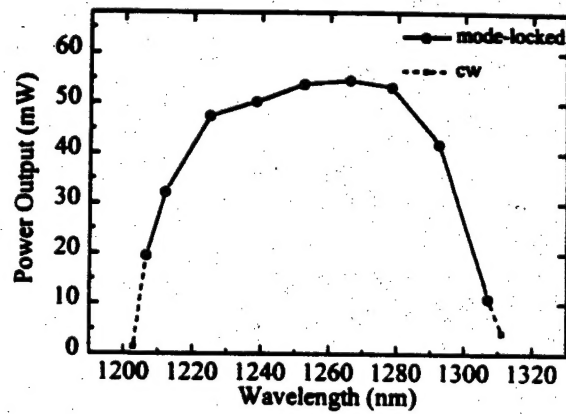


FIG. 3 Tunability curve of the Cr:forsterite laser with PbS QD doped glass saturable absorber (pump power=6.3W). The solid line indicates the wave-length region over which the laser was mode locked, whereas the dotted line indicates the wavelength region where the laser operates in plain cw.

We performed room temperature absorption measurements in several PbS quantum-dot (QD) doped glasses.² The quantum confinement effects were clearly observed in the well-resolved discrete absorption peaks spectrally shifted to shorter wavelengths from the bulk band edge. The PbS QD diameters for these samples were estimated from the energy shift of the absorption resonance. The particle diameters of 5.7, 6.8, 7.9, and 9.8 nm for the glasses with resonance peaks at 1206, 1386, 1563 and 1815 nm, respectively, were obtained (see Fig 4).

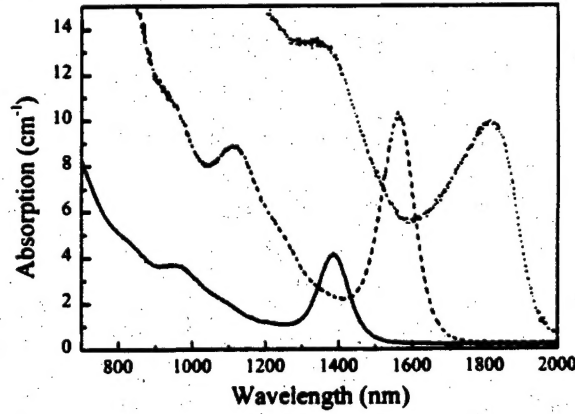


FIG. 4. Room temperature absorption spectra of glasses with PbS quantum dots. Diameters are: 6.6 (solid line), 7.5 (dashed line), and 9.3 nm (dotted line). The solid line corresponds to the sample used in this study.

The refractive index of these glasses at 1300 and 1550 nm are 1.513 and 1.510, respectively, measured with our prism coupler set-up. We performed single-beam absorption saturation measurements on PbS quantum-dot glass sample at room temperature (see Fig 5). The sample was 2 mm thick and had the absorption resonance at 1386 nm. For excitation pulses with 33 ps duration and wavelengths at $\lambda = 1.30 \mu\text{m}$ and $\lambda = 1.38 \mu\text{m}$ the saturation intensity was $I_{\text{Sat}} \approx 0.2 \text{ MW/cm}^2$ and $I_{\text{Sat}} \approx 0.4 \text{ MW/cm}^2$, respectively. This absorption saturation enables the modelocking described above.

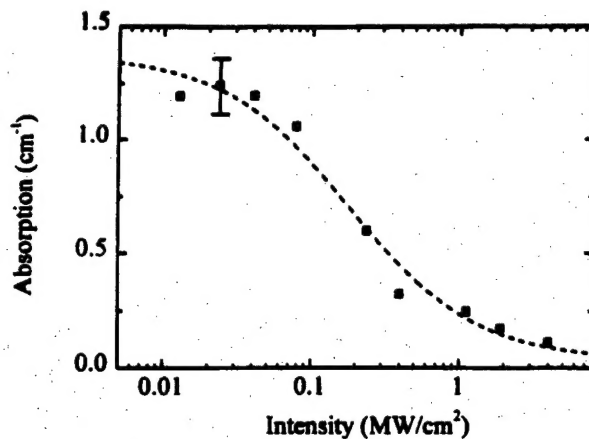


FIG. 5. Single beam absorption saturation data at 1.3 μm . The dashed line is a least-square fit to the model in Eq. (1). The error bars denote the uncertainty in the determination of the absorption due to the laser pulse fluctuations.

Finally, we developed a fabrication process for ion-exchanged waveguides in high quality passive glass. Our idea is to coat these waveguides with PbS-doped sol-gel thin films, which enables us to efficiently combine the excellent waveguiding properties of the ion-exchanged waveguides in passive glass and the non-linear properties of the PbS-doped thin films.

Optical properties of quantum dots by the sol-gel technique

Theory is predicting that three-dimensional quantum confinement should improve the properties of semiconductor (SC) devices. This enhancement has been seen in the absorption (nonlinear) properties until now, but not in the emission properties. The luminescence of SC quantum dots (QDs) is still dominated by trap related processes. The sol-gel fabricated QDs from UCLA have shown their improved quality by a drastic reduction of the photodarkening effects. They allow the incorporation of a high concentration of active material in the glass matrix with a relatively narrow dot size distribution.

At the University of Arizona, we studied the luminescence and the optical gain of the sol-gel derived CdS QDs. For the first time, we were able to report a significant optical gain in quantum dots pumped with nanosecond-pulse excitation. The gain persists up to room temperature, it has a broad spectral width, and its maximum value (more than 200 cm^{-1} at 10 K, about 30 cm^{-1} at room temperature) is located slightly below the absorption band edge. These features can be

surprising if we think at the delta function shaped density of states in zero dimensional SC, but it was confirmed by our theoretical calculations. The gain originates from the recombination of several excited levels between two and one electron-hole pairs states (i.e., biexciton to exciton). The luminescence dynamics of these CdS QDs was also studied. It showed that the intrinsic recombination occurs in picosecond (ps) time scale in our samples, and that the defect related emission happens in ns (nanosecond) time scale; i.e., much faster than in the conventional QDs which show the photodarkening effects. We also looked at thin films of CdS QDs dip-coated on a quartz plate. These samples had smaller dots and a bigger quantum confinement shift of the absorption.

Theory of ultrashort pulse propagation in quantum dots

On the theoretical side we have examined a wide range of semiconductor doped glasses for ultrashort pulse propagation studies and soliton formation for wavelengths below the half bandgap. By tailoring the linear dispersion soliton propagation should be possible. We have also continued studies of the modal properties of circular grating lasers and the properties at threshold.

Our recent theoretical efforts have been aimed at modeling ultrashort pulse propagation in quantum dot waveguides and fibers for wavelengths below the half bandgap. It has previously been shown that operating below the half bandgap has the combined virtues of low linear losses, though multi-photon absorption may become an issue, and enhanced ultrafast nonlinearities in the vicinity of two-photon resonance, offering unique opportunities for all-optical processing. In particular, the linear dispersion of the system can be combined with the nonlinearity to form optical solitons. However, in the vicinity of the half bandgap the quantum dots display significant nonlinear dispersion, resulting in optical shock formation, and destruction of the optical solitons. We have studied this nonlinear dispersion and evaluated its effects for a wide range of quantum dots. At first sight it would appear that optical solitons are therefore not possible, but further studies revealed that combining the nonlinear dispersion with higher-order linear dispersion can restore the optical solitons. This is a key result since methods to produce dispersion, such as using composite waveguides, invariably also produce higher-order dispersion,

which by itself would also destroy soliton formation, but the combination of the two effects can again allow solitons.

We have developed the above theoretical model and applied it to specific quantum dot systems. First, the linear and nonlinear optical properties of the quantum dot media are obtained using the Maxwell-Garnett theory for composite media, and these detailed material properties are used in ultrashort pulse propagation codes including linear dispersion, nonlinear self-phase modulation, and nonlinear dispersion. Our analysis shows that by suitable tailoring of the linear dispersion optical solitons can be formed. These theoretical studies open the doorway to propagation of space-time solitons, or optical bullets, in quantum dot waveguides.

Publications

1. P. T. Guerreiro, S. Ten, N. Borrelli, J. Butty, G. E. Jabbour, and N. Peyghambarian, "*PbS quantum-dot doped glasses as saturable absorbers for mode-locking of a Cr:forsterite laser*", Appl. Phys. Lett. **71**, 1595 (1997).
2. J. Butty, G. E. Jabbour, H. Tajalli, N. Peyghambarian, and N. Borrelli, "*Absorption saturation in PbS-quantum dots*", in proc. CLEO'96, Anaheim, June 2-7, 1996, p. 475.
3. M. Santagiustina and E. M. Wright, "*Suppression of third-order dispersion radiation in solid-state soliton lasers*," Opt. Lett. **20**, 2267 (1995).
4. P. T. Guerreiro, S. G. Lee, A. S. Rodrigues, Y. Z. Hu, E. M. Wright, N. Peyghambarian, S. I. Najafi, and J. Mackenzie, "*Femtosecond pulse propagation near a two-photon transition in a semiconductor quantum dot waveguide*," Opt. Lett. **21**, 659 (1996).
5. A. M. Dunlop, W. J. Firth, D. R. Heatley, and E. M. Wright, "*A generalized mean-field or master equation for nonlinear cavities with transverse effects*," Opt. Lett. **21**, 770 (1996).
6. A. M. Dunlop, W. J. Firth, and E. M. Wright, "*Master equation for spatio-temporal beam propagation and Kerr-lens mode-locking*," submitted to Opt. Commun.
7. M. Santagiustina and E. M. Wright, "*Linear and nonlinear dispersion in solid-state solitary-wave lasers*," contributed paper, OSA-IEEE XI Advanced Solid-State Lasers Topical Meeting, San Francisco, CA, February 1996.
8. A. M. Dunlop, W. J. Firth, and E. M. Wright, "*Generalized mean-field or master equation for nonlinear cavities with transverse effects*," contributed paper, CLEO 96, Anaheim, CA, June 1996.
9. P. T. Guerreiro, S. G. Lee, A. S. Rodrigues, Y. Z. Hu, E. M. Wright, S. I. Najafi, Y. H. Kao, J. Mackenzie, and N. Peyghambarian, "*Near two-photon resonance femtosecond pulse propagation in a CdS quantum dot waveguide*," contributed paper, CLEO 96, Anaheim, CA, June 1996.